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Multipole Mixtures in the Mössbauer Effect

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A theory of multipole mixtures applicable to resonance absorption is developed. The method used is an extension of Malus' law to include elliptically polarized multipole mixtures. The case of dipole-quadrupole mixtures is treated in detail as a means of measuring E2/M1 mixing ratios and checking time-reversal invariance in certain nuclei.

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## INTRODUCTION

Frauenfelder<sup>1</sup> and his co-workers have developed and applied the theory of elliptical polarization in the Mössbauer effect. In their paper (referred to hereafter as F) a method of complex vector parameterization was used to derive an expression for the transmission pattern,

$$I \Sigma = I I' \cos^2 \Theta \quad (1)$$

such that the factor  $I$ , which is proportional to the intensity of the emitted radiation, can be factored out on the right side of equation (1).

Then  $\Sigma$ , which is proportional to the absorption cross section, may be found explicitly in terms of the Euler angles  $\alpha, \beta$  for the oriented emitter and  $\alpha', \beta'$  for the oriented absorber. The angular factor,

$\cos^2 \Theta$ , is given in Table V of F for pure dipole and quadrupole radiation and for various values of  $M$  and  $M'$ , the changes in magnetic quantum number for emitter and absorber respectively. However, this method proved too complicated for a convenient treatment of multipole mixtures. It is the purpose of the present paper to develop the theory of multipole mixtures by a more direct method.

## THEORY

Let us begin with a modified form of equation (38) in F for the electric field vector,

$$E_{m2}(M) = a E_{m2}(M) + b e^{i\varphi} E_{m1}(M), \quad (2)$$

where  $a$  and  $b e^{i\varphi}$  are products of the appropriate Wigner  $3j$  symbols and reduced matrix elements  $\chi$ , as explained by F. We can choose  $a, b$

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and the relative phase of the nuclear matrix elements,  $\varphi$ , to be real numbers. The angular dependence is expressed by the vector

$$E_{\mu}(M) = e^{iM\gamma} (\hat{\eta}_1 e^{i\alpha} d_{1M}^{(L)}(\beta) \pm \hat{\eta}_{-1} e^{-i\alpha} d_{-1M}^{(L)}(\beta)) \quad (3)$$

where the upper signs refer to electric radiation and the lower signs to magnetic radiation. We shall be concerned with the case of most physical interest, namely, a mixture of magnetic dipole ( $L = 1$ ) and electric quadrupole ( $L = 2$ ) radiation. In the last equation,  $\gamma$  is the Euler angle measured about the axis of nuclear orientation,  $\beta$  is the angle between this axis and the axis of observation, while  $\alpha$  is the azimuthal angle measured about the axis of observation. The complex unit vectors are,

$$\hat{\eta}_{\pm 1} = \mp (\hat{i} \pm i\hat{j}) / \sqrt{2} \quad (4)$$

so that  $\hat{\eta}_{\mu}^* \cdot \hat{\eta}_{\nu} = \delta_{\mu\nu}$ , while the rotation matrix elements  $d_{\mu M}^{(L)}$  are given in Table II of F, reproduced here as Table I.

The quantity we are interested in is

$$\begin{aligned} I_Z &= |\mathcal{E}_{21}^*(M) \cdot \mathcal{E}'_{21}(M')|^2 \\ &= |aa' \mathcal{E}_2^*(M) \cdot \mathcal{E}'_2(M') + ab' e^{i\varphi'} \mathcal{E}_2^*(M) \cdot \mathcal{E}'_1(M') \\ &\quad + a'b e^{-i\varphi} \mathcal{E}_1^*(M) \cdot \mathcal{E}'_2(M') + bb' e^{-i(\varphi-\varphi')} \mathcal{E}_1^*(M) \cdot \mathcal{E}'_1(M')|^2. \end{aligned} \quad (5)$$

As a first step it is convenient to compute the complex numbers listed in Table II by using equation (3). From Table II and equation (5) with  $a$  or  $b$  set equal to zero, we can derive the formulas given in F. For example, with  $b = b' = 0$ , we may compute  $I_Z = |\mathcal{E}_2^*(\alpha) \cdot \mathcal{E}'_2(\pm 2)|^2$  which appears as the last entry in Table III. If in addition we let  $a = a'$ ,  $\alpha = \alpha'$ ,  $\beta = \beta'$ , and use the upper signs in this formula, we find the intensity squared and finally the intensity

$$I = \frac{1}{8} a^2 (4 \sin^2 \beta + \sin^2 2\beta) = \frac{1}{2} a^2 \sin^2 \beta (1 + \cos^2 \beta), \quad (6)$$

We may divide by this quantity to find

$$\Sigma = (I)^{-1} |\underline{\underline{E}}^* \cdot \underline{\underline{E}}'|^2 = I' \cos^2 \Theta \quad (7)$$

and divide again by  $I'$  to find

$$\cos^2 \Theta = (II')^{-1} |\underline{\underline{E}}^* \cdot \underline{\underline{E}}'|^2 \quad (8)$$

which appears as the first (or fourth) entry in Table V of F. We have omitted subscripts and arguments in equations (7) and (8) to indicate their general applicability even to multipole mixtures.

Before proceeding further, let us derive two auxiliary formulas which may be used to fill out Table II. The first is hardly more than a rearrangement of the quantities involved, as follows:

$$\underline{\underline{E}}_s^*(M) \cdot \underline{\underline{E}}_t^!(M') = [\underline{\underline{E}}_s(M) \cdot \underline{\underline{E}}_t^*(M')]^* = [\underline{\underline{E}}_t^*(M') \cdot \underline{\underline{E}}_s(M)]_x^* \quad (9)$$

where subscript  $x$  denotes the operation  $\alpha \leftrightarrow \alpha', \beta \leftrightarrow \beta', \gamma \leftrightarrow \gamma'$ .

This formula is convenient for finding such quantities as  $\underline{\underline{E}}_s^*(M) \cdot \underline{\underline{E}}_t^!(M')$  from  $\underline{\underline{E}}_t^*(M') \cdot \underline{\underline{E}}_s^!(M)$ . The second formula is

$$\underline{\underline{E}}_s^*(-M) \cdot \underline{\underline{E}}_t^!(\mp M') = (-1)^n [\underline{\underline{E}}_s^*(M) \cdot \underline{\underline{E}}_t^!(\mp M')]^* \quad (10)$$

where  $n = M + M' = (1 - \delta_{st})$  and  $\delta_{st}$  is the Kronecker delta. It is useful for obtaining  $\underline{\underline{E}}_s^*(-M) \cdot \underline{\underline{E}}_t^!(\mp M')$  from  $\underline{\underline{E}}_s^*(M) \cdot \underline{\underline{E}}_t^!(\mp M')$ . We may prove it by using equation (3) in equation (10) and equating the coefficients of the exponentials in the resulting expression. Thus we must show that

$$d_{1-M}^{(s)} d_{1 \mp M'}^{(t)} = (-1)^{M+M'} d_{-1M}^{(s)} d_{-1 \mp M'}^{(t)} \quad (11)$$

and

$$d_{1M}^{(s)} d_{1 \mp M'}^{(t)} = (-1)^{M+M'} d_{-1-M}^{(s)} d_{-1 \mp M'}^{(t)} \quad (12)$$

where we have ignored the symbol for the complex conjugate since the  $d_{\mu M}^{(L)}$  are real. These relations follow from the equation

$$d_{1-M}^{(L)} = (-1)^{1+M} d_{-1+M}^{(L)} \quad (13)$$

which can be obtained from an examination of Table I or, in the general case, from equation (4.19) of Rose<sup>2</sup>.

By using equations (9) and (10) to complete Table II we may now construct Table III from equation (5). In the last three entries of Table III one or both of the dipole components is missing, so that two of these formulas are only partial mixtures while the last is pure quadrupole. However, they are included for the sake of completeness. In order to fill out Table III we can employ two auxiliary transformations. To find  $I \Sigma(M, M')$  from  $I \Sigma(M', M)$ , we use

$$(\alpha, \beta, a, b, \varphi) \leftrightarrow (\alpha', \beta', a', b', \varphi'), \quad (14)$$

that is, exchange corresponding primed and unprimed quantities. To find  $I \Sigma(-M, -M')$  from  $I \Sigma(M, M')$ , we use

$$(a, a', b, b', \varphi, \varphi') \leftrightarrow (a, a', -b, -b', -\varphi, -\varphi') \quad (15)$$

with the Euler angles unchanged. These transformations may be proved by writing out equation (5) for the quantities involved and using equations (9) and (10) to transform one into the other. The transformation  $\gamma \leftrightarrow \gamma'$  is not included in equation (14) since this angle does not appear in the final result  $I \Sigma$ .

We may also compute the intensities given in Table IV from the first and third entries in Table III by using the upper signs and letting  $a = a'$ ,  $b = b'$ ,  $\alpha = \alpha'$  and  $\beta = \beta'$ . This gives us  $I_{21}(0)$  and

$I_{21}(+1)$  when we take the square root. If we use transformation (15) to find  $I_{21}(-1, \mp 1)$ , we may then obtain  $I_{21}(-1)$  by the same procedure. Similarly, from the last entry in Table III we obtain  $I_2(\mp 2)$  as in equation (6).

Finally, we note from equation (3) that electric and magnetic multipoles differ only in the sign of  $\hat{\eta}_1$ . As a result of this, Tables II, III, and IV are the same for a magnetic quadrupole-electric dipole mixture as for the electric quadrupole-magnetic dipole mixture we have been describing, although only the latter is of much physical interest.

#### EXPERIMENTAL POSSIBILITIES

Angular correlation measurements have been frequently used to determine the mixing ratio  $a/b$  and the relative phase  $\varphi$ <sup>3</sup>. At first<sup>4</sup> allowance was made for the possibility that the ratio is a complex number,  $a/(be^{i\varphi})$ . However, Lloyd<sup>5</sup> showed that the assumption of time-reversal invariance limits  $\varphi$  to the values 0 or  $\pi$ . Since the discovery of violations of the validity of parity conservation, attention has been turned toward experimental methods of checking time-reversal invariance also<sup>6-8</sup>. More recently, the Mössbauer effect has been proposed<sup>9</sup> as a technique for polarizing the daughter nucleus in an angular correlation experiment involving time-reversal and parity, and has been used<sup>10</sup> in a coincidence experiment to determine the E2/M1 mixing ratio of the 123-keV transition in Fe<sup>57</sup>.

The results of our paper might be used to determine  $a/b$  and  $\varphi$  for nuclei which show the Mössbauer effect<sup>11</sup> and are known to emit

mixed E2/M1 radiation<sup>12</sup>. Although only a limited number of such nuclei are known, and the Mössbauer effect requires the ground state to be the final state of a low energy ( $<150$ -keV) transition, still Zeeman experiments using only the Mössbauer effect can serve as a complement to the techniques described above. Of particular interest would be a more accurate check of time-reversal invariance for such nuclei.

### CONCLUSION

The theory of dipole-quadrupole mixtures has been presented in detail for emitter and absorber nuclei oriented in magnetic fields so that separated Zeeman lines appear. The method used is a traditional one since it amounts to an extension of Malus' law, discussed in most texts on optics. Both  $\underline{\mathcal{E}}$  (the "polarizer") and  $\underline{\mathcal{E}}'$  (the "analyzer") are projections of the electric vectors on the plane of observation, the  $\hat{n}_1, \hat{n}_2$  or  $\hat{i}, \hat{j}$  plane. Malus'  $\cos^2(\alpha - \alpha')$  law holds for the case of plane-polarized radiation,  $M = M' = 0$ . It is obtained by projecting one vector on the other, squaring the magnitude of the result and dividing by the intensities as in equation (8). We have extended this method to include elliptically polarized radiation for multipole mixtures and the pure multipoles which are special cases of these mixtures. Since equations (3) and (13) are perfectly general, the method may be extended to multipole mixtures of any order. Possible use of these results in experiments has also been briefly described.

Table I. Reduced rotation matrix elements,  $d_{\mu M}^{(L)}$ ,  
for dipole and quadrupole cases.

$\mu$	M	$d_{\mu M}^{(1)}$	$d_{\mu M}^{(2)}$
$\pm 1$	$\pm 2$	...	$\mp (2 \sin \beta + \sin 2\beta)/4$
$\pm 1$	$\pm 1$	$\cos^2 \beta / 2$	$(\cos \beta + \cos 2\beta)/2$
$\pm 1$	0	$\pm (1/\sqrt{2}) \sin \beta$	$\pm (3/8)^{1/2} \sin 2\beta$
$\pm 1$	$\mp 1$	$\sin^2 \beta / 2$	$(\cos \beta - \cos 2\beta)/2$
$\pm 1$	$\mp 2$	...	$\pm (2 \sin \beta - \sin 2\beta)/4$

Table IV. Intensities for quadrupole-dipole mixtures.

$$I_{21}(0) = (3/4) a^2 \sin^2 2\beta + b^2 \sin^2 \beta$$

$$I_{21}(\pm 1) = \frac{1}{2} [a^2 (\cos^2 \beta + \cos^2 2\beta) \pm 2ab \cos \varphi (\cos^2 \beta + \cos 2\beta) + b^2 (1 + \cos^2 \beta)]$$

$$I_2(\pm 2) = (1/8) a^2 (4 \sin^2 \beta + \sin^2 2\beta)$$



Table II. Complex numbers  $E_s^*(M) \cdot E_t'(M')$ .

$M$        $M'$        $E_s^*(M) \cdot E_t'(M')$

(a) Dipole radiation ( $s=t=1$ )

1	$\pm 1$	$\frac{1}{2} e^{-i(\gamma \mp \gamma')} [(1 \pm \cos \beta \cos \beta') \cos(\alpha - \alpha') - i(\cos \beta \pm \cos \beta') \sin(\alpha - \alpha')]$
$\pm 1$	0	$(1/\sqrt{2}) e^{\mp i\gamma} \sin \beta' [\pm \cos \beta \cos(\alpha - \alpha') - i \sin(\alpha - \alpha')]$
0	0	$\sin \beta \sin \beta' \cos(\alpha - \alpha')$

(b) Quadrupole radiation ( $s=t=2$ )

2	$\pm 2$	$\pm \frac{1}{2} e^{-i2(\gamma \mp \gamma')} \sin \beta \sin \beta' [(1 \pm \cos \beta \cos \beta') \cos(\alpha - \alpha') - i(\cos \beta \pm \cos \beta') \sin(\alpha - \alpha')]$
2	$\pm 1$	$-\frac{1}{2} e^{-i(2\gamma \mp \gamma')} \sin \beta [(\cos \beta' \pm \cos \beta \cos 2\beta') \cos(\alpha - \alpha') - i(\cos \beta \cos \beta' \pm \cos 2\beta') \sin(\alpha - \alpha')]$
$\pm 2$	0	$\mp (3/8)^{1/2} e^{\mp i2\gamma} \sin \beta \sin 2\beta' [\pm \cos \beta \cos(\alpha - \alpha') - i \sin(\alpha - \alpha')]$
1	$\pm 1$	$\frac{1}{2} e^{-i(\gamma \mp \gamma')} [(\cos \beta \cos \beta' \pm \cos 2\beta \cos 2\beta') \cos(\alpha - \alpha') - i(\cos \beta' \cos 2\beta \pm \cos \beta \cos 2\beta') \sin(\alpha - \alpha')]$
$\pm 1$	0	$(3/8)^{1/2} e^{\mp i\gamma} \sin 2\beta' [\pm \cos 2\beta \cos(\alpha - \alpha') - i \cos \beta \sin(\alpha - \alpha')]$
0	0	$\frac{3}{4} \sin 2\beta \sin 2\beta' \cos(\alpha - \alpha')$

(c) Cross terms ( $s=1, t=2$ )

1	$\pm 2$	$\mp \frac{1}{2} e^{-i(\gamma \mp 2\gamma')} \sin \beta' [(\cos \beta \pm \cos \beta') \cos(\alpha - \alpha') - i(1 \pm \cos \beta \cos \beta') \sin(\alpha - \alpha')]$
1	$\pm 1$	$\frac{1}{2} e^{-i(\gamma \mp \gamma')} [(\cos \beta \cos \beta' \pm \cos 2\beta') \cos(\alpha - \alpha') - i(\cos \beta' \pm \cos \beta \cos 2\beta') \sin(\alpha - \alpha')]$
$\pm 1$	0	$(3/8)^{1/2} e^{\mp i\gamma} \sin 2\beta' [\cos(\alpha - \alpha') \mp i \cos \beta \sin(\alpha - \alpha')]$
0	$\pm 2$	$\mp (1/\sqrt{2}) e^{\pm i2\gamma'} \sin \beta \sin \beta' [\cos(\alpha - \alpha') \mp i \cos \beta' \sin(\alpha - \alpha')]$
0	$\pm 1$	$(1/\sqrt{2}) e^{\pm i\gamma'} \sin \beta [\cos \beta' \cos(\alpha - \alpha') \mp i \cos 2\beta' \sin(\alpha - \alpha')]$
0	0	$-i(\sqrt{3}/2) \sin \beta \sin 2\beta' \sin(\alpha - \alpha')$

Table III. Transmission factors  $I_{\Sigma} = |\underline{\varepsilon}_{2,1}(M) \cdot \underline{\varepsilon}_{2,1}'(M')|^2$

M	M'	$I_{\Sigma}$
0	0	$\sin^2 \beta \sin^2 \beta'$ $\times \left\{ \left[ (3aa' \cos \beta \cos \beta' + b b' \cos(\varphi - \varphi')) \cos(\alpha - \alpha') + \sqrt{3} (a b' \sin \varphi' \cos \beta - b a' \sin \varphi \cos \beta') \sin(\alpha - \alpha') \right]^2 \right.$ $\left. + \left[ \sqrt{3} (a b' \cos \varphi' \cos \beta + b a' \cos \varphi \cos \beta') \sin(\alpha - \alpha') + b b' \sin(\varphi - \varphi') \cos(\alpha - \alpha') \right]^2 \right\}$
$\pm 1$	0	$\frac{1}{2} \sin^2 \beta'$ $\times \left\{ \left[ (\sqrt{3} a a' \cos \beta' \cos 2\beta + a b' \cos \varphi' \cos \beta + \sqrt{3} b a' \cos \beta' \cos \varphi \pm b b' \cos(\varphi - \varphi') \cos \beta) \cos(\alpha - \alpha') \right. \right.$ $\left. \pm (a b' \sin \varphi' \cos 2\beta - \sqrt{3} b a' \cos \beta' \sin \varphi \cos \beta \mp b b' \sin(\varphi - \varphi')) \sin(\alpha - \alpha') \right]^2$ $+ \left[ (\sqrt{3} a a' \cos \beta' \cos \beta \pm a b' \cos \varphi' \cos 2\beta \pm \sqrt{3} b a' \cos \beta' \cos \beta \cos \varphi + b b' \cos(\varphi - \varphi')) \sin(\alpha - \alpha') \right.$ $\left. + (-a b' \sin \varphi' \cos \beta + \sqrt{3} b a' \cos \beta' \sin \varphi \pm b b' \sin(\varphi - \varphi') \cos \beta) \cos(\alpha - \alpha') \right]^2 \right\}$
1	$\pm 1$	$\frac{1}{4} \left\{ \left[ a a' (\cos \beta \cos \beta' \pm \cos 2\beta \cos 2\beta') + a b' \cos \varphi' (\cos 2\beta \pm \cos \beta \cos \beta') \right. \right.$ $\left. + b a' \cos \varphi (\cos \beta \cos \beta' \pm \cos 2\beta') + b b' \cos(\varphi - \varphi') (1 \pm \cos \beta \cos \beta') \right] \cos(\alpha - \alpha')$ $+ \left[ a b' (\cos \beta \pm \cos \beta' \cos 2\beta) - b a' \sin \varphi (\cos \beta' \pm \cos \beta \cos 2\beta') - b b' \sin(\varphi - \varphi') (\cos \beta \pm \cos \beta') \right] \sin(\alpha - \alpha') \Big\}^2$ $+ \frac{1}{4} \left\{ \left[ a a' (\cos \beta' \cos 2\beta \pm \cos \beta \cos 2\beta') + a b' \cos \varphi' (\cos \beta \pm \cos \beta' \cos 2\beta) \right. \right.$ $\left. + b a' \cos \varphi (\cos \beta' \pm \cos \beta \cos 2\beta') + b b' \cos(\varphi - \varphi') (\cos \beta \pm \cos \beta') \right] \sin(\alpha - \alpha')$ $+ \left[ -a b' \sin \varphi' (\cos 2\beta \pm \cos \beta \cos \beta') + b a' \sin \varphi (\cos \beta \cos \beta' \pm \cos 2\beta') + b b' \sin(\varphi - \varphi') (1 \pm \cos \beta \cos \beta') \right]$ $\times \cos(\alpha - \alpha') \Big\}^2$
$\pm 2$	0	$\frac{1}{2} \sin^2 \beta \sin^2 \beta'$ $\times \left\{ \left[ (\pm \sqrt{3} a a' \cos \beta \cos \beta' + a b' \cos \varphi') \cos(\alpha - \alpha') \pm a b' \sin \varphi' \cos \beta \sin(\alpha - \alpha') \right]^2 \right.$ $\left. + \left[ (\sqrt{3} a a' \cos \beta' \pm a b' \cos \varphi' \cos \beta) \sin(\alpha - \alpha') - a b' \sin \varphi' \cos(\alpha - \alpha') \right]^2 \right\}$
2	$\pm 1$	$\frac{1}{4} \sin^2 \beta$ $\times \left\{ \left[ (a a' \{ \cos \beta' \pm \cos \beta \cos 2\beta' \} + a b' \cos \varphi' \{ \cos \beta \pm \cos \beta' \}) \cos(\alpha - \alpha') \right. \right.$ $\left. + a b' \sin \varphi' (1 \pm \cos \beta \cos \beta') \sin(\alpha - \alpha') \right]^2$ $+ \left[ (a a' \{ \cos \beta \cos \beta' \pm \cos 2\beta' \} + a b' \cos \varphi' \{ 1 \pm \cos \beta \cos \beta' \}) \sin(\alpha - \alpha') \right.$ $\left. - a b' \sin \varphi' (\cos \beta \pm \cos \beta') \cos(\alpha - \alpha') \right]^2 \Big\}$
2	$\pm 2$	$\frac{1}{4} a^2 a'^2 \sin^2 \beta \sin^2 \beta' [(\cos \beta \pm \cos \beta')^2 + \sin^2 \beta \sin^2 \beta' \cos^2(\alpha - \alpha')]$

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